Non-Universal Power Law of the "Hall Scattering Rate" in a Single-Layer Cuprate $Bi_2Sr_{2-x}La_xCuO_6$

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In-plane resistivity, Hall coefficient, and magnetoresistance (MR) are measured in a series of high-quality $Bi_2Sr_{2-x}La_xCuO_6$ crystals with various carrier concentrations, from underdope to overdope. It is found that the temperature dependence of the Hall angle obeys a power law T^α with α systematically decreasing with increasing doping, which questions the universality of the Fermi-liquid-like T^2 dependence of the "Hall scattering rate". The systematics of MR indicates an increasing role of spin scattering in underdoped samples.

The peculiar normal-state properties of high-T_c cuprates are generally considered to be the keys to elucidate the high-T_c mechanism, since they give us a clue to clarify the nature of the strongly-correlated electronic state of the cuprates. To understand the underlying electronic state of the cuprates, it is desirable to study the normal state at low temperatures [1] and to chase the systematic evolution thereof with carrier concentration [2]. Therefore, particularly useful system for the normal-state study is such a system where T_c is relatively low and where the the carrier concentration can be changed in a wide range from underdope to overdope.

A typical study in which a wide temperature window for the normal-state is desirable is the measurement of the temperature dependence of the scattering time. For example, the observation of the T-linear resistivity from 10 K to 700 K in $Bi_2Sr_2CuO_6$ (Bi-2201) [3] had a strong impact, because it clearly demonstrated the dominance of the electron-electron interaction in the scattering mechanism. In high-T_c cuprates, it has been discussed that the charge transport is governed by two different scattering times with different temperature dependences [4–6]; according to this "two-scattering-time" model, in-plane resistivity ρ_{ab} is governed by the transport scattering time τ_{tr} ($\sim T^{-1}$) and the Hall angle θ_H is governed by the "Hall scattering time" τ_H ($\sim T^{-2}$). However, this idea of "scattering-time separation" has not yet gained a complete consensus and there are other approaches to understand the unusual normal-state transport properties [7,8]. Therefore, it would be useful to establish the temperature and doping range in which the T^2 behavior of the Hall scattering rate τ_H^{-1} is observed.

There are two widely-known cuprate systems which satisfy the requirements of the relatively low T_c and the availability of a wide doping range; $La_{2-x}Sr_xCuO_4$ (LSCO) system and Bi-2201 system. Bi-2201 has not been as intensively studied as LSCO, mostly because of the difficulty in obtaining high-quality single crystals. A number of problems have been known for Bi-2201 crystals: (a) the transport properties of B-2201 are quite non-reproducible even among crystals of nominally the same composition [9–11]; (b) the residual resistivity of the in-plane resistivity ρ_{ab} is usually large (the smallest value reported to date is 70 $\mu\Omega$cm [3,11]), as opposed to LSCO, $YBa_2Cu_3O_7$ (YBCO), or $Bi_2Sr_2CaCu_2O_8$ (Bi-2212), where the residual resistivity in high-quality crystals is negligibly small; and (c) the temperature dependence of the Hall coefficient is weak and thus the Hall angle θ_H does not obey the T^2 law [10,11], while $\theta_H \sim T^2$ has been almost universally observed in other cuprates [6]. On the other hand, the Bi-2201 system has very attractive characteristics: it has been known for Bi-2201 that the carrier concentration can be widely changed by partially replacing Sr with La (to underdope) or Bi with Pb (to overdope) [12]; at optimum doping ($Bi_2Sr_{2-x}La_xCuO_6$ with $x \simeq 0.4$), the maximum T_c is about 30 K [12,13], which is lower than the maximum T_c of LSCO. Therefore, if single crystals of sufficiently high quality are grown, this system would present an ideal stage for the systematic study of the normal-state properties down to lower temperatures than in other cuprates.

In this Letter, we report that it is possible to obtain a series of high-quality Bi-2201 crystals and show that in those high-quality crystals the normal-state transport properties display behaviors which are in good accord with other cuprates; for example, in the underdoped region, ρ_{ab} shows a downward deviation from the T-linear behavior at a certain temperature which decreases with increasing doping and, in the overdoped region, T dependence of ρ_{ab} changes to T^n with $n > 1$. What is new in this system is that the Hall angle indeed obeys a power law T^α with $\alpha \simeq 2$, but the power α shows a systematic decrease towards smaller values as the carrier concentration is increased. This finding of a systematic change of α in a relatively-simple single-layer cuprate system poses a serious question to the universality of the "Fermi-liquid like" T^2 behavior of the Hall scattering rate τ_H^{-1} [6]. Another notable finding is that the ratio of the longitudinal magnetoresistance (MR) to the transverse MR increases systematically with decreasing carrier concentration, in-

dicating that a spin contribution to the MR gradually becomes significant as the sample becomes more underdoped.

The single crystals of $Bi_2Sr_{2-x}La_xCuO_6$ (BSLCO) are grown using a floating-zone technique in 1 atmosphere of flowing oxygen. It is known that pure Bi-2201 is an overdoped system [12]. Since the La substitution to the Sr site reduces the number of holes, increasing La doping brings the system from overdoped region to underdoped region. We found that the La substitution results in a growth of crystals of better morphology compared to the pure Bi-2201. The actual La concentrations in the crystals are determined by the inductively-coupled plasma spectrometry (ICP) technique. Here we report crystals with x=0.24, 0.30, 0.44, 0.57, and 0.66, for which the zero-resistance T_c is 24, 30, 33, 28.5, and 20 K, respectively. The inset to Fig. 1 shows the zero-resistance T_c (and the mid-point T_c) as a function of x. Apparently, $x \simeq 0.4$ corresponds to the optimum doping, which is consistent with previous reports on BSLCO [12,13]. The onset of the Meissner effect (measured with a commercially available Quantum Design SQUID magnetometer) for the optimally-doped crystals is 33 K. To our knowledge, this optimum T_c is the highest value ever reported for Bi-2201 or BSLCO system. The ICP analysis found that our crystals are Bi-deficient by about 0.2, while Sr+La is almost stoichiometric; for example, the x=0.44 sample has the composition of $Bi_{1.8}Sr_{1.57}La_{0.44}CuO_{6+\delta}$.

ical size of $2 \times 1 \times 0.015$ mm^3. The thickness of the crystals are accurately determined by measuring the weight of the sample with 0.1 μg resolution; therefore, the uncertainty in determining the magnitude of the resistivity is less than $\pm 5\%$. The crystals are annealed at 400°C for 30 minutes in flowing oxygen upon firing silver epoxy. We use a standard six-terminal method for simultaneous magnetoresistance (MR) and R_H measurements, in which the data are taken with an ac technique in the sweeping magnetic field at fixed temperatures. The temperature is very carefully controlled and stabilized using both a capacitance sensor and a Cernox resistance sensor to avoid systematic temperature deviations with magnetic fields. The stability of the temperature during the MR and R_H measurements is within a few mK.

Figure 1 shows the temperature dependence of ρ_{ab} for the five x values in zero field. Clearly, both the magnitude of ρ_{ab} and its slope show a systematic decrease with increasing carrier concentration (decreasing x). We found that it is only at the optimum doping that ρ_{ab} shows a perfect T-linear behavior. Figure 2 shows the temperature dependence of the slope $d\rho_{ab}/dT$; only the x=0.44 sample shows a constant $d\rho_{ab}/dT$, which corresponds to the T-linear behavior, in a wide temperature range (from 300 K to 120 K). We note that a fitting of the ρ_{ab} data of the x=0.44 sample to $\rho_{ab}=\rho_0 + AT$ gives the residual resistivity ρ_0 of only 25 $\mu\Omega$cm, which, to our knowledge, is the smallest value ever reported for pure Bi-2201 or BSLCO.
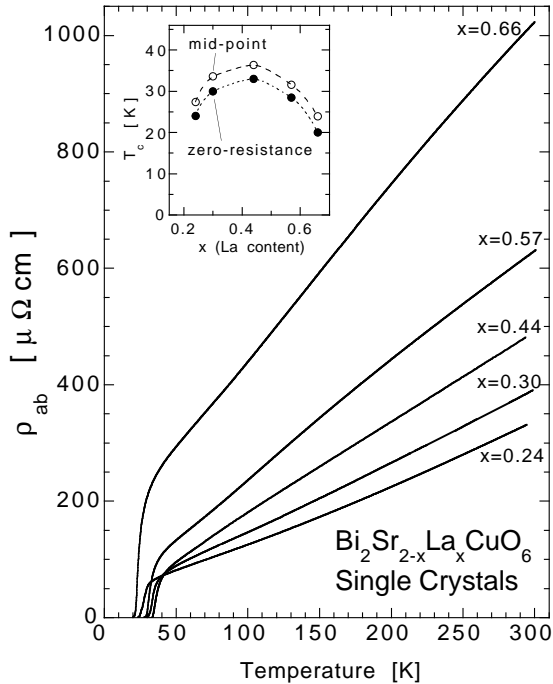

FIG. 1. T dependence of ρ_{ab} of the BSLCO crystals. Inset: T_c as a function of La content x. The dashed lines are guide to the eyes.

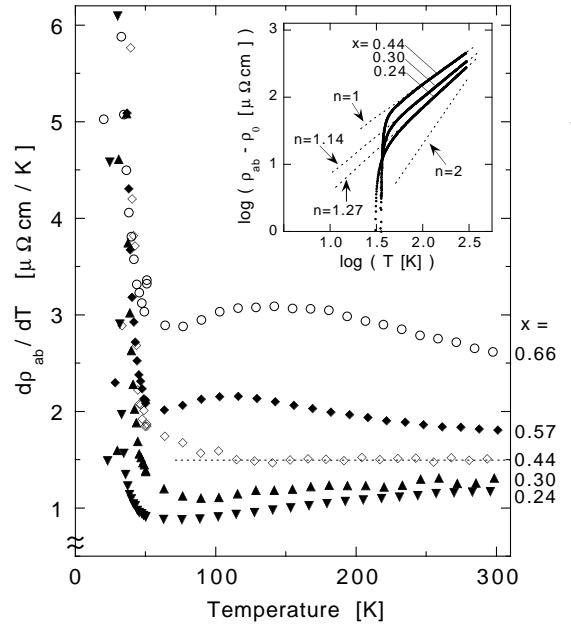The crystals are cut into a rectangular shape with typ-

FIG. 2. T dependence of the slope $d\rho_{ab}/dT$. The dashed line represents a constant slope (T-linear behavior). Inset: Plot of $\log(\rho_{ab}-\rho_0)$ vs $\log T$ for three x values to show the power-law dependence $\rho_{ab}=\rho_0 + AT^n$, which is represented by dotted lines.

The underdoped samples (x=0.57 and 0.66) show a rather complex temperature dependence in ρ_{ab}, which is similar to that of underdoped YBCO [14]; the behavior in underdoped YBCO is characterized by a downward deviation from the T-linear dependence and the presence of a maximum in $d\rho_{ab}/dT$. Such a behavior has been correlated to the opening of a pseudogap [14]. The broad maximum in $d\rho_{ab}/dT$ moves to higher temperature as the carrier concentration is decreased, which agrees with the conjecture that the pseudogap opens at higher temperature in more underdoped samples. On the other hand, the slope $d\rho_{ab}/dT$ of the overdoped samples (x=0.30 and 0.24) monotonically decreases with decreasing temperature. This reflects the fact that ρ_{ab} of the overdoped samples behaves as ρ_{ab}=$\rho_0 + AT^n$ with n larger than 1, a behavior reported in the overdoped samples of LSCO [15] and $Tl_2Ba_2CuO_{6+\delta}$ (Tl-2201) [16]. The inset to Fig. 2 shows that $\rho_{ab}(T)$ for x=0.30 and 0.24 can actually be described by the power law with n=1.14 and 1.27, respectively. Therefore, in both the underdoped and overdoped regions, the behavior of $\rho_{ab}(T)$ shows an evolution which can be considered to be "standard" for high-T_c cuprates. This observation indicates that Bi-2201 is not an exceptional system but rather is a promising system for the systematic study of the normal-state transport properties.
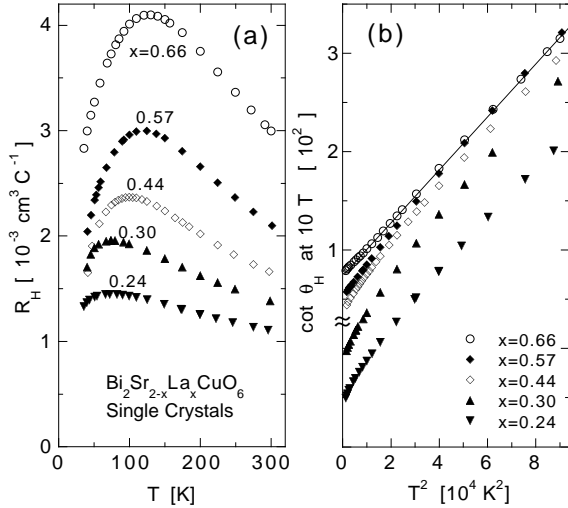
holds only in this most underdoped sample. We found that $\cot\theta_H$ for other x values obey a power law T^α with α smaller than 2, which is shown in Figs. 4 (a) - (d). The best powers are 1.85, 1.70, 1.65, and 1.60, for x =0.57, 0.44, 0.30, and 0.24, respectively. In all the panels of Fig. 4, the data are very well fitted with straight lines. Therefore, the power-law temperature dependence of the Hall scattering rate τ_H^{-1} holds for every doping in BSLCO, but the power α shows a systematic decrease with increasing carrier concentration [17]. A particularly intriguing fact here is that $\cot\theta_H$ of the optimally-doped sample changes as $T^{1.70}$, not as T^2, while ρ_{ab} shows a good T-linear behavior. This might mean that the "Fermi-liquid like" behavior of $\tau_H^{-1}\sim T^2$ (which has been proposed to be the characteristic of spinons [5]) may not be a generic feature of the optimally-doped cuprates.
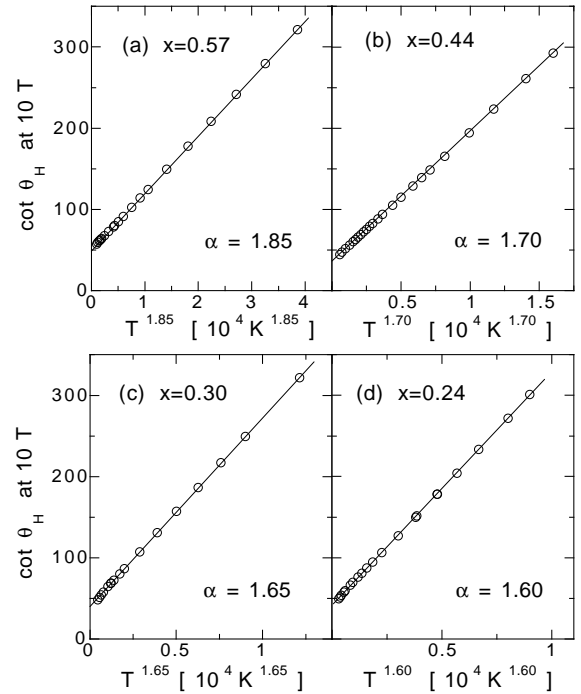

FIG. 4. Plots of $\cot\theta_H$ vs T^α for the data of (a) x=0.57, (b) x=0.44, (c) x=0.30, and (d) x=0.24. The solid lines are fits to the data.

FIG. 3. (a) T dependence of R_H of the BSLCO crystals. (b) Plot of $\cot\theta_H$ vs T^2 (the data for x=0.30 and 0.24 are shifted down by 50 and 100, respectively, to avoid congestion). The solid line is a fit to the x=0.66 data.

Figure 3(a) shows the temperature dependence of R_H for the five samples. A clear evolution of R_H with x is observed; the change in the magnitude of R_H at 300 K suggests that the carrier concentration is actually reduced roughly by a factor of 3 upon increasing x from 0.24 to 0.66. Figure 3(b) shows the plot of $\cot\theta_H$ ($=\rho_{xx}/\rho_{xy}$) vs T^2. Only the data for x=0.66 can be fitted with a straight line in this plot, indicating that the T^2 law of $\cot\theta_H$

Figure 5 shows the transverse and longitudinal MR of four of the samples, x=0.24, 0.44, 0.57, and 0.66. One may immediately notice a trend that the relative magnitude of the longitudinal MR compared to the transverse MR increases with increasing x. (We omit the result of x=0.30 due to the limited space; the behavior of this sample fits well into the trend.) Since it is expected that the longitudinal MR mostly comes from a spin contribution (while the transverse MR consists of both an orbital and the spin contribution), our result suggests that the role of spins in the transport becomes increasingly significant as the sample becomes more underdoped.
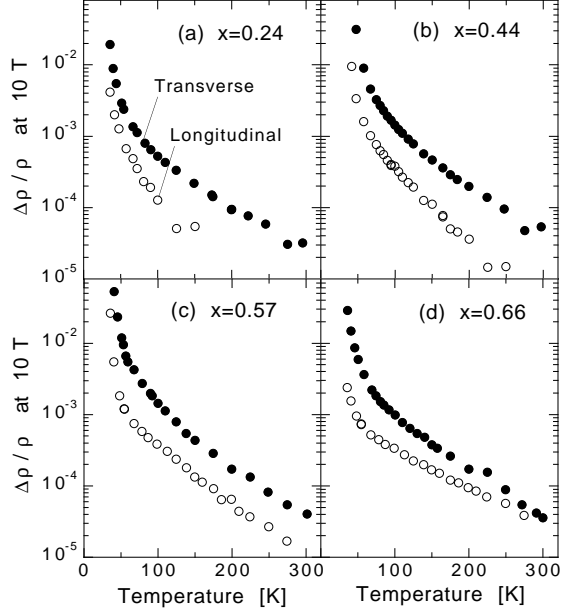
FIG. 5. Transverse MR (solid circles) and longitudinal MR (open circles) for (a) x=0.24, (b) x=0.44, (c) x=0.57, and (d) x=0.66.

In passing, let us briefly mention our analysis to see if any scaling is applicable to the MR data. Firstly, we found that the classical Kohler's rule $\Delta\rho/\rho \sim (H/\rho)^2$ is strongly violated, as in other cuprates [18,19]. We also found that the "modified Kohler's rule" $\Delta\rho/\rho \sim (\cot\theta_H)^{-2}$ is not very well applicable to our data; this is different from the results for LSCO and YBCO [18,19] and might be related to the fact that $\cot\theta_H$ does not behave as T^2 except for the x=0.66 sample. The details of the MR analysis will be published elsewhere.

The above results tell us altogether that the systematics of the power law of the scattering rates ($\tau_{tr}^{-1} \sim T^n$ and $\tau_H^{-1} \sim T^\alpha$) needs to be reconsidered. In particular, τ_H^{-1} shows a tendency that the power α becomes systematically smaller with increasing doping in the whole doping range studied. This observation, combined with the systematic change of the longitudinal MR, might suggest that the T^2 law of τ_H^{-1} is observable only when the spin contribution to the charge transport is strong. This in turn suggests that in Bi-2201 the role of spin degrees of freedom is already weakened at optimum doping, which might be the reason for the relatively low T_c of this system. Interestingly, another single-layer cuprate Tl-2201 shows a good T^2 dependence of τ_H^{-1} at optimum doping [20] and Tl-2201 has the maximum T_c of 85 K. Finally, because of the relatively wide temperature range in which the normal-state transport properties can be studied, BSLCO system offers an ideal stage for the detailed study of the systematic evolution of the scattering times as well as other normal-state properties.

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